COMPARISON OF ACCELERATOR-BASED WITH REACTOR-BASED NUCLEAR WASTE TRANSMUTATION SCHEMES

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ABSTRACT

An overview of the most significant studies in the last 35 years of partitioning and transmutation of commercial light water reactor spent fuel is given. Recent Accelerator-based Transmutation of Waste (ATW) systems are compared with liquid-fuel thermal reactor systems that accomplish the same objectives. If no long-lived fission products (e.g., ^{99}Tc and ^{129}I) are to be burned, under ideal circumstances the neutron balance in an ATW system becomes identical to that for a thermal reactor system. However, such a reactor would need extraordinarily rapid removal of internally-generated fission products to remain critical at equilibrium without enriched feed. The accelerator beam thus has two main purposes (1) the burning of long-lived fission products that could not be burned in a comparable reactor's margin (2) a relaxing of on-line chemical processing requirements without which a reactor-based system cannot maintain criticality. Fast systems would require a parallel, thermal ATW system for long-lived fission product transmutation. The actinide-burning part of a thermal ATW system is compared with the Advanced Liquid Metal Reactor (ALMR) using the well-known Pigford-Choi model. It is shown that the ATW produces superior inventory reduction factors for any near-term time scale.

Introduction

The scientific uncertainties in planning the first US nuclear waste repository, located at Yucca Mountain, Nevada, are large. Additionally, geological disposal of nuclear waste, an endeavor that is "unprecedented in human history" (Otis, 1991) requires us to make predictions about human behavior for hundreds or thousands of years into the future (Erikson, 1994). Because of these reasons and growing political opposition, its opening has been delayed until at least the year 2010 (GAO, 1993). Further, the expected costs have grown from about $1 billion to $25.7 billion (Keeney and Winterfeldt, 1994). It is estimated that there are 60,000 tonnes of spent fuel (33,000 MW-days burnup) currently available to be sent to this repository, which is limited by statute to hold at most 70,000 tonnes of spent fuel (DOE, 1984; DOE, 1986). The amount of spent fuel in storage will likely double by the time the repository is opened.

Transmutation of nuclear waste offers the possibility of increasing the public acceptability and decreasing the cost of high level waste disposal. Most proposed systems involve fissioning some or all of the actinides, which would have the benefit of extending US energy resources. There is potentially great

revenue from the sale of electricity, but probably not enough to offset completely the cost of fuel reprocessing (Keeny et al., 1977; Croff et al., 1980). If the wholesale price of electricity were to increase significantly in the future because of, e. g., the environmental costs of fossil fuel combustion being added to the price of fossil fuel as a tax, this balance could change.

The proposal to use the Advanced Liquid Metal Reactor (ALMR) for actinide burning has raised significant interest (Wade, 1994). The main problem with this or other schemes using a fast spectrum is the very large core inventory, resulting in very long time of operation required to obtain a significant degree of transmutation per unit inventory. Because of this, a commitment to continue some form of the ALMR for many hundreds to thousands of years would be necessary (Pigford and Choi, 1991). Actinide burning by itself would make only a negligible reduction in the risk to the public from contaminated water from the repository, because the species most likely to migrate from the repository are the water-soluble long-lived fission products (LLFPs), especially ^{99}Tc, ^{135}Cs and ^{129}I (Pigford, 1991).

Only by relatively complete elimination of the higher actinides and LLFPs can the time scale for the storage possibly be reduced. Because of advances in accelerator technology (Lawrence et al., 1991) in recent years, it appears that accelerator-based transmutation of waste (ATW) systems have become more feasible (Powell et al., 1981; Bowman et al., 1992; Venneri et al., 1993; Beard et al., 1994a). An ATW system offers the advantage of transmuting some of the LLFPs. A small repository or engineered storage facility could then hold a large number of reactor-years of the remaining waste, without the legacy of leaving the most hazardous long-lived components of the waste unattended for millennia.

In this paper we begin with a brief but hopefully useful description of the spent fuel nuclear waste problem and a review of the transmutation concepts that have been proposed over the years. We make the case that accelerator-based transmutation schemes offer some unique advantages versus other schemes, most of which are based on actinide recycle in LWR's or fast reactors. We show that actinide-burning ATW systems can be thought of as the combination of an energy-producing actinide-burning thermal reactor and an energy-consuming LLFP-burning target/blanket system. The thermal reactor component resembles the old Oak Ridge Molten Salt Breeder Reactor concept, which we describe in some detail. It has an advantage over LWR's in that it allows a much higher thermal neutron flux. Depending on the chemical processing system capabilities that are coupled with this reactor, it may act as a net source or sink of neutrons. The results of scoping studies are used to show the role of the accelerator in a system that burns both actinides and LLFPS. We then use the model of Pigford and Choi (1991) to compare a system using a fast reactor to transmute the actinides to one that uses the thermal reactor implied by the ATW system. It is shown that a faster inventory reduction is achieved with the thermal reactor.

Waste Characterization

The LWR spent fuel nuclides of interest are discussed in many references, for instance Benedict, et al., (1981), Hebel, et al. (1978), or Binney, et al. (1990). The major categories (long- and short-lived fission products, activation products and actinides) each possesses specific characteristics. The fission products and activation products are characterized by beta emission and with the exception of a few nuclides, possess significantly shorter half-lives than the transuranics of concern. The actinides exhibit alpha-decay and some spontaneous fission. While a number of transuranics possess short half-lives, they typically have long-lived daughter products which characterizes their chains by extremely long half-lives.

The main risk from a geological repository is usually stated as the risk from contaminated groundwater (Pigford, 1991). Of the species in LWR spent fuel, it is often concluded that only a few are capable of reaching the water table and have the potential to reach the accessible environment (Ramspott et al., 1992, Hirschfelder et al., 1991). These species are ^{99}Tc, ^{129}I, ^{135}Cs, ^{237}Np, ^{242}Pu ,^{238}U ,^{234}U. Because of solubility in water, lack of soil retention, and long half live only a few species are listed as the ones likely

to pose risk from groundwater contamination. It cannot be stated with certainty that this list will not change with further study. Transuranic burning addresses only the species ^{237}Np and ^{242}Pu on the list. Most ATW systems address the burning of ^{99}Tc, ^{129}I and sometimes ^{135}Cs.

Croff et al. (1991) discuss the effects of actinide burning on the in-repository inventory relative to the then-existing EPA standards.[1] Figures 1 and 2 are taken from Croff et al. (1991), and can be used as a qualitative tool for understanding the benefits and limitations of transmutation on repository performance. Figure 1 shows a calculation of the repository inventory (in Ci) normalized to its cumulative release limit (in Ci). This calculation, because it is based on the EPA standard includes consideration of transport, uptake and uncertainties in the analysis. The dotted area represents the range of acceptability according to the standard. The higher the limit, the lower the probability of exceeding it must be. If the ratio is less than unity, the repository has met the standard *a priori*. As can be seen in the figure, the inventory exceeds the standard by many orders of magnitude for at least 10^5 years. The repository thus depends on the *geological* isolation of the waste from the environment for that period of time. A lower curve depicts the contents of repository waste after actinide burning. The waste approaches the range of acceptability at 1000 years. Assumed reprocessing losses are given in the upper right hand corner of the figure. The lowest curve indicates the inventory *sans* actinides.

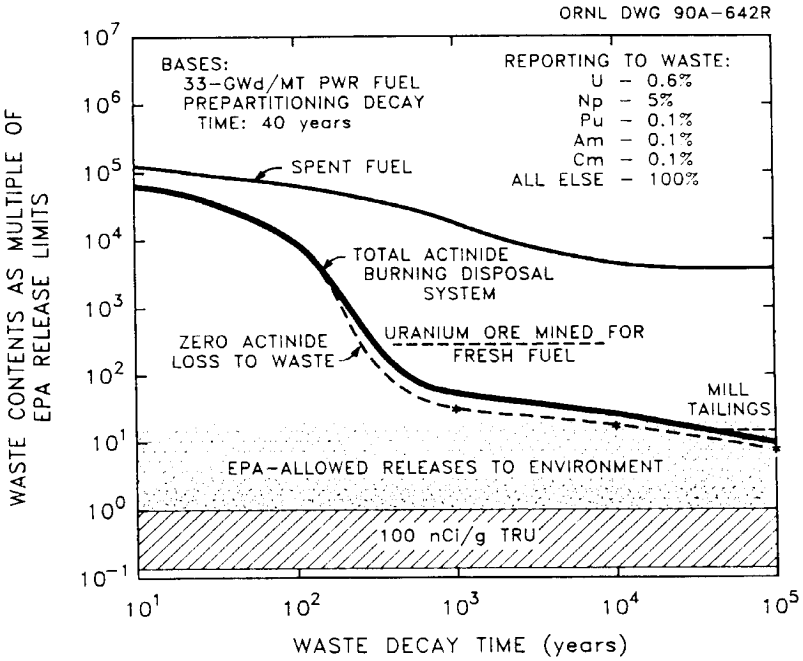

Fig. 1. Radionuclide inventory of repository relative to EPA disposal standard with and without actinide burning (taken from Croff et al. (1991))

Figure 2 shows the principal nuclides contributing to the inventory after actinide burning. By this measure, the most hazardous materials at early times less than ~600 years are ^{90}Sr and ^{137}Cs, at times from ~600 years to ~30000 years ^{14}C, ^{59}Ni and ^{240}Pu, and at longer times ^{59}Ni, ^{93}Zr and ^{99}Tc. Transmutation of the ^{99}Tc and ^{129}I removes the two species that are most likely to contaminate groundwater. Several species are not normally listed as posing a risk for groundwater contamination but do posses long half-lives, e. g., ^{93}Zr, ^{59}Ni and ^{14}C. Because of difficulty in separating and transmuting these isotopes, they may represent a floor below which further reductions in risk from the transmutation of other immobile species are fruitless. Complete elimination of the need for some sort of repository or long-term storage facility would be difficult to achieve because of these species. However, reprocessing of the spent fuel opens the possibility to increase repository safety by creating new waste forms that are optimized for the different radionuclides that are not transmuted (Pigford et al., 1983; Ramspott et al., 1992). In example, an

[1]A review of the standards is underway by a National Academy of Sciences panel that is due for completion by the end of 1994.

optimized waste form for ^{14}C could be an insoluble carbonate or carbide. The Cs and Sr separated during processing can be stored in a monitored surface facility for hundreds of years, until the ^{137}Cs and ^{90}Sr have decayed; the remaining Cs would either then be placed in a repository or the ^{135}Cs content could be transmuted. If the repository capacity is limited by heat loading, the decreased volumetric heat generation would allow a much larger amount of reprocessing waste to be placed in a given area. This facility would also hold the actinide residue from reprocessing unless much cleaner reprocessing technology can be developed.

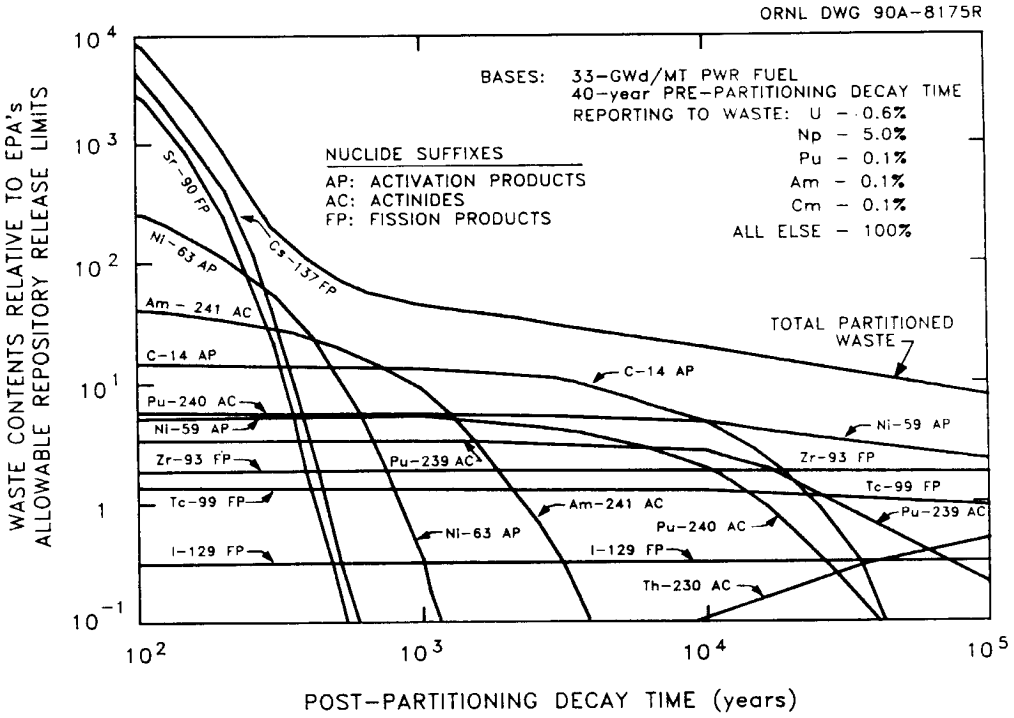

Fig. 2. Principle radionuclide contributors to repository inventory relative to EPA disposal standard (taken from Croff, et al. (1991))

Since actinide transmutation relies on the ability to remove the nuclides from the rest of the waste with a high degree of purity, the chemical processing (partitioning) of the high-level waste is crucial. Croff et al., (1980) described the various chemical processes required to remove the actinides from the high-level waste and to include them in fuel fabrication for reactor recycle. Using the chemical processing techniques available at the time (1979), 99.75% of the actinides could be removed from the high-level waste stream with 0.1% being lost to waste in reprocessing, and 0.15% being lost during fuel fabrication. A more recent study (Binney et al., 1990) utilized recent technological developments in the separation processes. In this approach, spent fuel is first dissolved in a nitric acid solution. The plutonium and uranium are removed using the aqueous-based PUREX process (McKay et al., 1989). The TRUEX process (Schultz and Horwitz, 1987; 1988) is utilized to remove the remaining transuranics. Croff et al. (1991) mention newer processing technologies that have been demonstrated on a laboratory scale which may be effective for further removing of actinides from solid waste streams. Non-aqueous pyroprocessing reprocessing technology is currently being developed at Argonne National Laboratory (ANL) as part of the Integral Fast Reactor (IFR) program (Ramspott et al., 1992, Wade, 1994, Thompson, 1990). It is metal-based and can be adapted to separation of transuranics from LWR discharge spent fuel. Processing losses are similar to aqueous technology.

Other non-aqueous front-end and back-end processing systems are possible. The zirconium cladding and most of the uranium contained in the spent fuel can be removed by the RENUW process, a modification of the dry fluoride volatility (Johnson, 1992) process. The LWR spent fuel assemblies might be treated first by an eddy-current or by a plasma torch process to melt and remove the metallic zirconium cladding from the oxide spent fuel. The oxide rubble would then be subjected to fluoridation which converts the large

amount of uranium to UF_6. The highly volatile UF_6 would be readily removed and the remaining fluorinated actinides, including solid PuF_4 and the fission products, would then be fed into the transmutation system as a liquid salt. The separation of the fission products from the actinide fluorides (back-end) is handled on line by a liquid centrifuge system (Bowman, 1992).

As is discussed in the Ramspott et al. study, transmutation schemes usually do not address the uranium isotopes, and no one has made any plans for disposal of the stockpiled depleted uranium that has collected near the US government's enrichment facilities. If transmutation were used to eliminate all the species on this list except uranium, there would be the need to study how to dispose of or transmute this species.

Reactor Transmutation Overview

The earliest transmutation studies (Steinberg et al., 1958; Gregory and Steinberg, 1967), investigated the possibility of transmuting the toxic waste isotopes, ^{85}Kr, ^{90}Sr, and ^{137}Cs in a thermal fission reactor. The amounts of these nuclides disposed of in a geologic repository could be reduced by a factor of 1000 with neutron fluxes of 10^{16} neutrons/(cm^2-sec). The high flux required is due to the small capture cross sections of these isotopes. However, the disadvantages such reactors possess (short core-life, high decay heat, further waste production) make this approach impractical.

These studies were followed by an examination of how to enhance the waste management process by reducing the long-lived toxicity of high-level radioactive wastes (Claiborne, 1972; Claiborne, 1975). The long-lived toxicity (> 1000 years) of high-level radioactive waste could be reduced to about 5% of that of pitchblende by the removal of 99.99% of plutonium, 99.9% of uranium, americium, and curium, and 95% of neptunium, which would be recycled back into the nuclear fuel cycle. The toxicity could be reduced by only a factor of approximately four at decay times of 1000 years because the actinides are not truly eliminated, but are maintained at an equilibrium level (the quantities of actinides at which the production from capture in the fuel is equal to the destruction of the actinides due to fission).

The first significant studies (Beaman, 1975; Beaman and Aitken, 1976) of using fast reactors for actinide recycle were of liquid-metal fast breeder reactors (LMFBR's). In these studies actinides from three 1000 MWe BWR's and one 1000 MWe LMFBR were recycled into one LMFBR by loading them into special fuel pins and placing them in a concentric ring in the LMFBR core. Beaman found that the actinide equilibrium concentrations could be obtained in nine cycles as compared to the twenty cycles required for a PWR recycling the actinide wastes from a single PWR (as determined by Claiborne, 1972). Consequently the fast spectrum of an LMFBR is superior for use in actinide recycle. Murphy et al. (1979) determined that the specific reactivity of the actinides was directly proportional to the average neutron energy, and therefore actinide recycle was best accomplished using specially designed hard-spectrum reactors in which the majority of power is produced from the fission of the waste actinides. These fast reactors also build up their in-core inventory of higher actinides to equilibrium values which are higher than the initial core loading.

The actinide equilibrium build-up amounts (IAEA, 1982) are factors by which the quantity of actinide feed must be multiplied by in order to obtain the equilibrium amounts that will be discharged at the end-of-life from a reactor employing recycle. They range from 1.2 to 60 depending on the isotope and the type of reactor and are given in Table 1. Although the equilibrium amounts are greater than the normal discharge, the actual inventory of actinides is reduced since this is the amount of waste which must be dealt with at the end of the reactor's lifetime (approximately 30 years) as opposed to the end of a single batch (approximately 3 years). The IAEA concluded that while technically feasible, the marginal decrease in the long-term radiological hazard did not justify actinide recycle. The study also identified possible difficulties in implementation of actinide recycle including the incorporation of the actinides into the existing fuel fabrication process and proving that reactor safety and reliability are not compromised.

Table 1: Build-Up Factors of Principal Waste Actinides (IAEA, 1982). These factors are the amount which the quantity of actinide feed must be multiplied by in order to obtain the equilibrium amounts which will be discharged at the end-of-life from the reactor employing recycle.

	PWR-U	PWR-Pu	LMFBR
^{237}Np	2.1	1.9	2.4
^{238}Pu	2.4	2.0	unavailable
^{241}Am	1.2	1.2	2.4
^{243}Am	1.5	1.3	3.1
^{244}Cm	9.4	5.1	15.0
^{245}Cm	14	5.8	58

Two large studies, one by Oak Ridge (Croff et al., 1980) and the other by Hanford (Binney et al., 1990) assessed the feasibility of transmuting the actinides after separation was achieved. The Oak Ridge study concluded that transmutation of the actinides did not yield significant enough benefits with regard to toxicity or waste reduction to warrant implementation. Alternatively, the Binney et al. study concluded that partitioning and transmutation could be beneficial by making the design and operation of a repository system easier due to the reduced heat generation rates. The plutonium would be fissioned while cesium and strontium could either be transmuted or sequestered.

The proposal to use the Advanced Liquid Metal Reactor (ALMR) for actinide burning has raised significant interest. The proposed program requires reprocessing enough LWR spent fuel to fuel the initial core and the first two reloads of the ALMR. Subsequent reload cores would be produced by reprocessing ALMR spent fuels. System make-up is from reprocessed LWR spent fuel. The emphasis is on complete burning of all transuranics, and thus the ALMR may appear to qualify as a transmutation scheme.

The Integral Fast Reactor (IFR) concept may overcome proliferation objections (Keeny et al., 1977) by providing technical barriers to diversion of transuranic materials (Chang and Till, 1991; Wade, 1994). The recycle of the higher actinides in this solid-fueled system is via a pyrometallurgical technology that is unable to achieve a fission product decontamination factor greater than about 1000; thus, the transuranic product is self-protecting as a result of the gamma radiation from (especially) the lanthanides. The chemistry further cannot produce a plutonium product that is not at least 25% contaminated with uranium; the plutonium is always mixed with the minor actinides which provide isotopic contamination and substantial decay heat and neutron radioactivity. The pyrometallurgical recycle technology and the pyrophoric nature of the metallic fuel form and recycle product dictate that the recycle processes be conducted remotely in a heavily shielded, argon-inerted cell; the limited accessibility allows implementation of substantial physical control measures.

As a result of the development of the ALMR, advances in reprocessing technology and growing public resistance to a repository, the authors of an important Oak Ridge paper (Croff et al., 1991) drew some new conclusions about the incentives for actinide burning. They stated that although "a commitment to undertake or deploy actinide burning is premature until a better basis for a decision is in hand" that it "should be regarded as a technological option that may have benefits for waste management and the energy security of the United States."

Because many experts had expressed opinions similar to this one, a study was conducted to determine the impact that a decade of changes has had on partitioning and transmutation (P-T) as a waste management option (Ramspott et al., 1992). Their main conclusion was that there had been no changes that would

affect the main economic conclusion of Croff et al.'s 1980 study. In particular, they focused on the ALMR and the effects that it would have on the total inventory of actinides that would exist in the country with and without its use. Because of the low rate at which a fast reactor burns actinides versus its own inventory, a commitment to continue some form of the ALMR for many hundreds to thousands of years would be necessary (Pigford and Choi, 1991). In addition, the ultimate reduction in actinide inventory in the repository waste would make only a negligible reduction in the risk to the public from contaminated groundwater near the repository. The waste volume in a repository would remain roughly constant, because they did not include sequestering of short-lived isotopes in their calculations. Their main objection to P-T was that it may require or encourage a delay in the opening of the first US repository.

The overall negative tone of the Ramspott et al. study towards P-T was punctuated by some positive statements. They noted that "the most powerful argument for P-T is certainty. Removal of radioactive isotopes provides an ability to predict future effects that is unchallengeable. If radionuclides do not exist, they cannot cause an effect". In reducing risk "P-T does assist by reducing the impact of human intrusion scenarios". The study was also careful to point out that the long-term risk to the public is dominated by migration of LLFPs, rather than actinides, in the groundwater.

Accelerator Transmutation Overview

There are some real limits to transmutation in conventional solid-fueled reactor systems, e. g., they require a resonance absorber (typically ^{238}U) to provide a negative doppler coefficient. Unless an alternative material is substituted (e. g. erbium or tungsten), this will always lead to the production of more higher actinides. Fast reactor systems have the problem of large inventory and positive sodium void coefficient for actinide burning and a neutron energy spectrum that gives too low of an LLFP capture rate per unit inventory. Thermal reactors in general are inappropriate for burning LLFP species because they have an inadequate reactivity margin and too low of a flux level. Addition of a neutron source somewhere in a thermal transmutation scheme can help to overcome these problems without the shortfalls of a fast system.

Early transmutation studies considered large accelerators to be too far off in the future to be a factor, but there has been an evolution in the understanding of the accelerator and spallation target source. While thermonuclear fusion appears to be the most attractive source, it has not been proven as a near-term technology, and consequently is not a viable option at present. Spallation neutron sources afford significant advantages over existing high-flux reactors due to their ability to generate a much greater effective flux (Lovesey and Stirling, 1984). The spallation source is a promising neutron source for transmutation applications, not only due to its low heat deposition, but also because it produces only few fission products, and no transuranics unless fissionable material is incorporated into the target design.

The spallation process involves the interaction of a high-energy particle with a nucleus of an atom, thus ejecting nucleons. Protons of between 1 and 2 GeV are the preferred projectiles (Fraser, 1977). The residual nucleus is left in an excited state, decaying by evaporation. The total yield is dependent upon the energy and type of incident particle, and the material with which the incident particle collides. Figure 3 displays the experimentally-determined spallation neutron yield from protons on various materials as a function of energy. As can be seen, the neutron production is a linear function of energy, with the greatest yields at the highest energies. Consequently, in order to obtain a sufficient neutron yield to make a spallation source advantageous compared to other options, energies of approximately 1 GeV are required. This eliminates the use of neutrons and heavy ions as the incident particles due to the problems associated with producing these particles in the required energy range. Protons, deuterons, and tritons, however, can all be accelerated to sufficient energies, and trade-offs must be considered in selecting among these as the most appropriate particle. While deuterons and tritons produce a greater neutron yield for a given energy, they tend to cause greater activation problems in the accelerator structure.

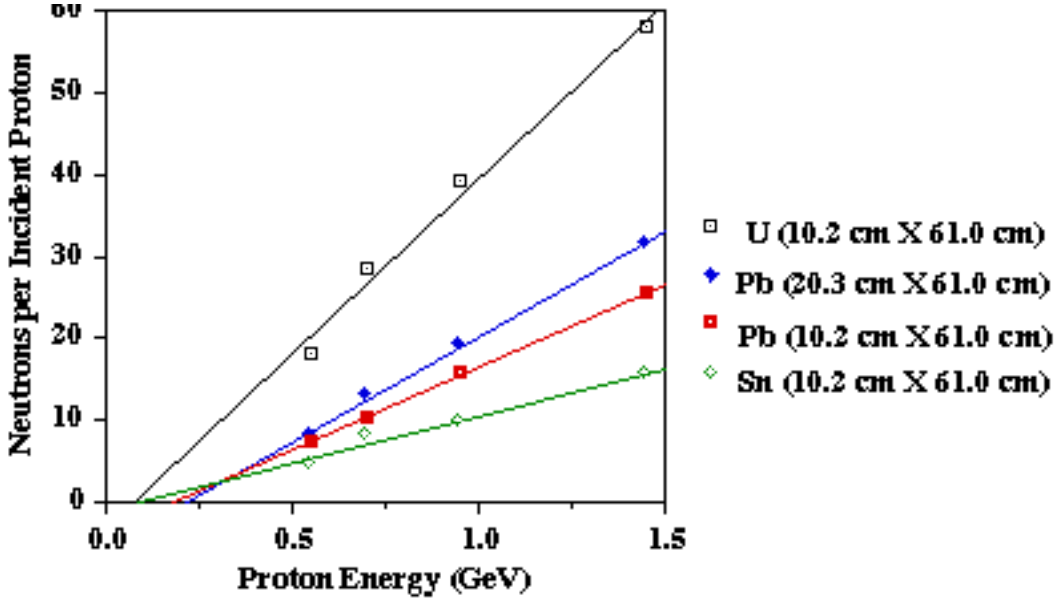

Fig. 3. Neutron yield from various spallation targets as a function of incident proton beam energy (from Fraser, 1965).

Achieving the required energies at sufficient beam currents for a spallation source to be feasible for waste transmutation makes the selection of the accelerator system a vital issue (Lawrence et al., 1991). In a study conducted by Brookhaven and Los Alamos National Laboratories on accelerator production of tritium (Billen et al., 1993) the linear accelerator was identified as the only type of accelerator capable of accelerating a steady, continuous beam of over 100 mA. Linear accelerators are superior to cyclotrons in that they provide the most efficient means of energy conversion and have the lowest beam loss factor. In a detailed analysis, the report identifies the highest-powered accelerator which can be developed using existing technology, a 1600 MeV proton, 250 mA linear accelerator, which will be used as the reference accelerator in this study.

The spallation process produces an extensive number of nuclides which are not normally produced in fission reactors. In addition, the distribution of nuclides produced is substantially different than that produced through fission. As an example, the LAHET-calculated (Prael and Lichtenstein, 1989) spallation product distribution produced by bombardment of lead with 1600-MeV protons in shown in Figure 4. This code has been repeatedly validated against experimental data (Brun et al., 1993). The spallation process produces nuclides which span the entire range of elements up to the mass (and even slightly greater) of the target material. However, the greatest production occurs in a pronounced peak near the target material's mass, because usually only a few nucleons are ejected in any single reaction. Hence, the nuclides produced in this peak will typically be dominant contributors to the waste stream, and these nuclides depend upon the target material. Materials such as lead and bismuth produce a number of long-lived isotopes (^{194}Hg, ^{202}Pb, ^{205}Pb, ^{208}Bi) in significant quantities, while tungsten and tantalum only have a single long-lived isotope in their primary production peak (^{182}Hf). A second peak occurs in the intermediate-mass nuclides due to the generation of high-energy fission. As the target mass increases, this peak will also increase and can become a major contributor to the waste stream. Consequently, the final waste stream resulting from the use of a spallation target will depend upon the detailed design and the materials present in the target, as well as the energy and current of accelerated particles to which it is exposed. These species have very low β decay energies and are insoluble in groundwater meaning that they would have probably no effect on repository performance.

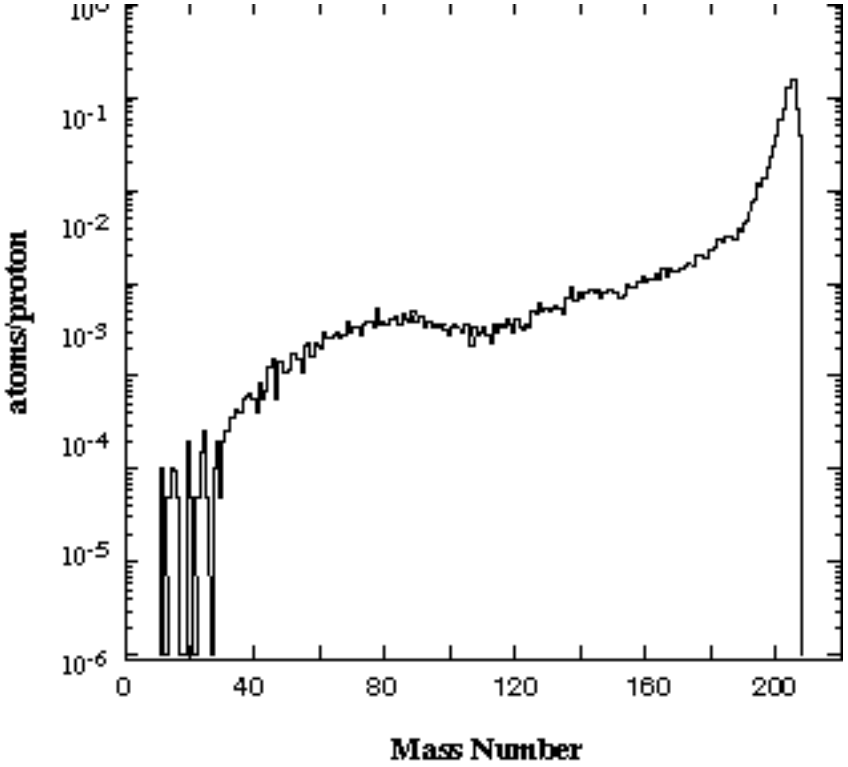

Fig. 4. Calculated spallation product distribution produced by 1600-MeV protons on a lead target. The curve was calculated using the code LAHET (Prael and Lichtenstein, 1989).

A variety of spallation neutron sources have been designed and built for various purposes. Table 2 lists the major spallation neutron source facilities in use and gives some of their characteristics. However, all of these sources were constructed for research purposes, and consequently the accelerators are of insufficient energy, beam current, and duty factor for use in transmutation. Also, their targets are not designed to withstand the high power densities which would be generated in a high-energy, high-beam-current system.

Table 2: Existing Spallation Sources (Gregory and Steinberg, 1967; Fraser and Bartholomew, 1983). All the listed sources are for research purposes.

Facility	Type	Beam	Energy (MeV)	Current (μA)	Target
LAMPF-WNR	LINAC	proton	800	100	W
KENS	synch.	proton	500	1.9	U^{238}
SNS	synch.	proton	800	200	U
TRIUMF	cyclotron	proton	500	< 100	Pb

The first significant investigation of using a spallation system for high-level waste transmutation was the LAFER/APEX project conducted at Brookhaven National Laboratory (Steinberg et al., 1979). The initial basis of this project was the development of a spallation breeder for use in generating ^{239}Pu or ^{233}U from ^{238}U or ^{232}Th, respectively. The system consisted of a 1.5 GeV proton accelerator operating at 300 mA impinging upon a flowing liquid lead target, with a blanket of fertile material (either ^{238}U or ^{232}Th) surrounding the target. In one alternative system for transmutation the ^{90}Sr and ^{137}Cs were placed in the spallation breeder system. The effective half-lives of these isotopes could be reduced by a factor of ten versus recycling these wastes back into fission reactors. No details were provided as to how these nuclides were to be placed in the system. Isotopic separation of ^{137}Cs would be required in order to keep ^{133}Cs and ^{135}Cs from dominating the neutron absorption process.

The ATW concepts of Los Alamos in recent years have emphasized complete burning of the higher actinides in a thermal spectrum after separation from spent fuel. The LLFPs ^{99}Tc and ^{129}I are burned as well. Some of these systems have proposed burning other isotopes, e. g., ^{135}Cs either totally or partially. The two most discussed are the graphite-moderated molten salt system (Bowman et al., 1992; Venneri et al., 1993) and the heavy-water moderated aqueous slurry system (Beard et al., 1994a). Figure 5 shows a generic conceptual design for an ATW target/blanket assembly that burns both the higher actinides and LLFPs. The target would consist of a material that produces neutrons from spallation reactions. A significant amount of high-Z material, such as lead, is required to maximize neutron yield. The blanket contains a moderated heterogeneous lattice, using a flowing fluid which contains the actinides. Fission heat is carried away by convection and the fluid is cooled in an external heat exchanger. There are separate regions in the blanket where the LLFPs are transmuted.

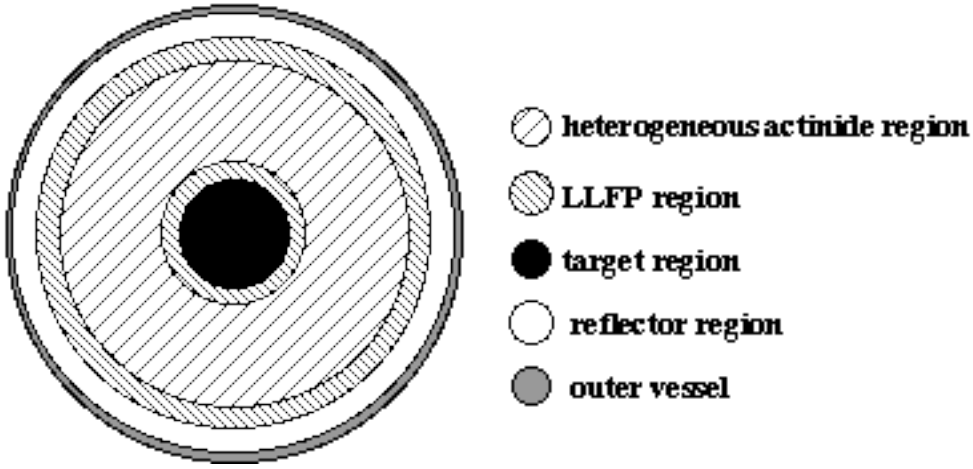

Fig. 5. The conceptual ATW system combining actinide transmutation and LLFP transmutation in the same blanket.

Fission products continually are produced in the fuel and must be removed by processing, or else they will become a very significant neutron poison for the system. The LLFPs produced by fission are removed by the chemical system and stored for later transmutation. Another chemistry system must be present to remove LLFP transmutation products (e. g., ^{100}Ru from transmutation of ^{99}Tc) .

In the molten salt system, the fuel is in the form of a fluoride salt (~100's g/l AcF_4), dissolved in a molten salt carrier whose composition is 50% ^{7}LiF / 50% $^{9}BeF_2$. The carrier's melting point and operating temperature are 500 °C and 650 °C, respectively. The molten salt flows through channels of circular cross section bored in a hexagonal pattern directly into the graphite moderator lattice. In the selected design, the power density is about 400 MW/m^3 in the fuel. The parasitic neutron absorber ^{135}Xe is removed continuously by a helium gas stripping loop. The target, centrally located, is of liquid lead.

Design details of the aqueous system are available in Beard et al. (1994a). It will not be discussed further although much of the analysis can be applied to this system as well. The neutron energy spectra in the two systems are highly thermal, but the parasitic capture rates are slightly higher in the aqueous system, where the high-pressure tubing and tungsten target are significant sinks for thermal neutrons. The capture-to-fission ratio for the fuel is a little higher because the graphite is not as effective a moderator as heavy water. Also, the important fission product ^{135}Xe is soluble enough in the water to remain trapped until it absorbs a neutron. In the molten salt system, there is some absorption in graphite. The ^{135}Xe is stripped out effectively and does not cause neutron loss. The overall conclusions are that neutron multiplication factors of 12-15 are possible in either system, which will burn all the higher actinides in LWR spent fuel, along with the ^{99}Tc, ^{129}I in the spent fuel and all ^{99}Tc, ^{129}I produced by the higher actinide fission.

The ATW system as shown in the above figure can be broken into two separate subsystems. There is a reactor to burn the actinides (that may also burn some fission products), and there is a separate accelerator-driven target/blanket assembly to burn LLFPs. The fluid-fueled actinide-burning reactor is shown in the left hand side of **Figure 6**. This reactor concept resembles the fluid-fueled concepts that were studied in the 1950's and 60's (Lane et al., 1958). Again, fission product concentrations must be kept to a minimum by continuous removal, requiring the use of fluid fuels. The prompt negative temperature coefficient of the fluid fuels eliminates the need for a resonance absorber. The same heterogeneous lattice is used. The flowing fuel is cooled in an external heat exchanger, and a chemical processing system is present to remove fission products. The remaining ATW system, shown in the right hand side, would burn LLFPs only.

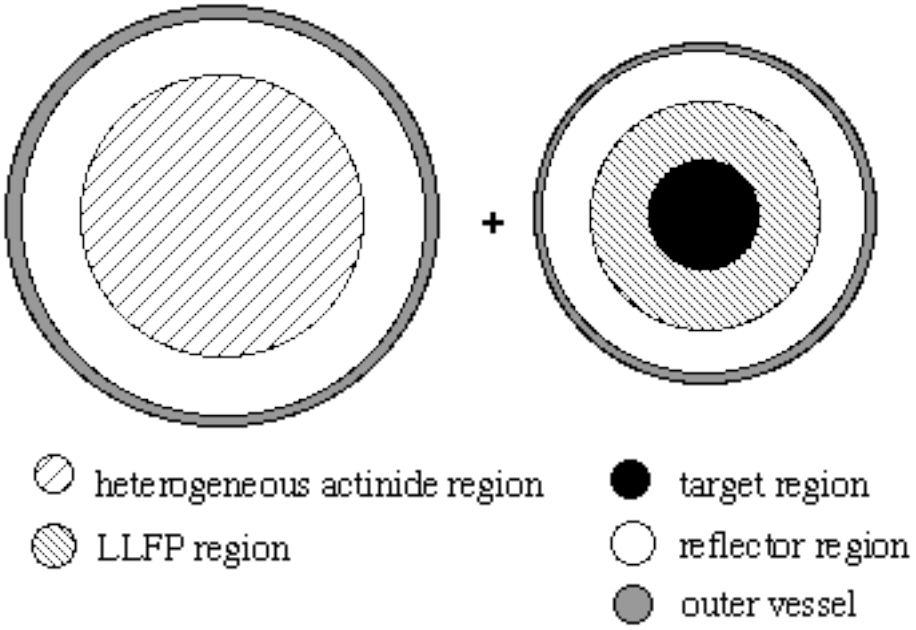

Fig. 6. (left) An actinide burning reactor that is based on the same heterogeneous lattice as the ATW system. (right) the remaining LLFP-burning ATW blanket.

According to our calculations, described in some detail in the next sections, the reactor may be able to form a critical configuration throughout its lifetime, completely transmuting the LWR waste feed into fission products. There may also be a small reactivity margin available to transmute some LLFPs in these reactors. The neutron balance in a real system will, of course, be a function of design detail. For instance, in general, the higher the power density, the lower the *equilibrium* capture/fission ratio for the actinides. In practical systems it may prove difficult to prevent flux dilution by holdup of the fuel in external components, e. g., heat exchangers or hold-up tanks. The neutron balance in the reactor component is also a sensitive function of the removal rates of the fission products if a thermal spectrum is used. Whether rapid and reliable fission product removal can be attained is discussed later in some detail.

There are two reasons for breaking the ATW system into two subsystems. The main reason, for the purpose of this paper, is to provide a conceptual tool for understanding the detailed energy and mass balances in these systems. Such a breakdown reveals the two true roles of the accelerator in the transmutation scheme, i. e., (1) to provide extra neutrons to burn LLFPs, and (2) to burn entrained fission products that may not be effectively removed by chemical processing systems. Additionally, we can begin to make trade studies on the reactor component of the system by itself. The use of this conceptual tool depends on the acceptance of the idea that the two configurations are nearly equivalent in terms of neutron balance. According to our calculational experience, the main difference between the configurations is that the leakage is somewhat higher when there are two subsystems rather than one.

The second reason to decompose ATW into these components may be a practical one. There may be some operational and/or safety advantages to literally building ATW as two side-by-side subsystems. One subsystem would be a critical or slightly subcritical (accelerator-driven) one to transmute actinides. The other would be non-multiplying and would transmute LLFPs with an accelerator beam and spallation source. The separate systems may allow designers to choose two different neutron spectra in the two systems. Maintenance ease and reliability may be higher with two subsystems rather than one.

The Reactor Component

The molten salt and aqueous homogeneous reactors were extensively studied and tested during the 1950's and 60's. The safety advantages of these systems are well documented (Lane et al., 1958; Shimazu, 1977). The strong negative temperature coefficient provided by the liquid fuels makes these systems passively safe against excursions. If xenon is continuously purged from the fuel, control rods may be unnecessary except for shutdown purposes.

A complete neutron balance for the burning of actinides and LLFPs from LWR spent fuel under the assumption of several different neutron energy spectra is given in the excellent paper by Salvatores et al. (1994). They performed the complete bookkeeping so as the find the result of all possible capture vs. fission vs. decay branches. If the neutrons liberated per fission are known for each species in each type of spectrum, the net neutron balance for complete burning can be calculated. This means that no actinides are put into a repository or disposed of in any other way than fission. Each feed isotope (species i, having an atom fraction in the feed N_i) will undergo an average number of capture reactions $<\alpha_i>$ before absorbing another neutron causing fission. The only endpoint for each nucleus is fission, which will liberate a weighted average number of neutrons, $<\nu_i>$. The net number of neutrons consumed to burn a feed isotope is therefore $D_i = <\alpha_i> + 1 - <\nu_i>$. The net number of neutrons to burn a feed stream in a given spectrum is then $D = \Sigma\ D_i N_i$. In fast spectra the value of D for a LWR discharge stream is very negative, indicating that transmutation of all transuranic species without an external neutron source is possible. There would even be a surplus of neutrons for further transmutation of LLFPs, if necessary. For thermal spectra, they give D values nearer to zero, but still negative. The exception occurs upon addition of fuel highly enriched in ^{235}U, where the surplus of neutrons is much greater, and the value of D becomes more negative.

In most of our work, and in the work presented in this paper, we assume equilibrium in performing our neutron balances. To find the relationship between the D values of Salvatores and "equilibrium values" of the number of neutrons liberated per absorption in the fuel, η_{eq}, one should imagine a transmutation system at steady state with a fixed spectrum. By definition, neither the composition nor the inventory of the actinides in the system is changing with time. Over a time Δt, one mole of actinides is introduced into the system. Over this same period of time one mole undergoes fission. By Salvatores et al.'s reasoning, the net number of neutrons emitted from the reactions in the system is (1 - D) moles. By our method this quantity is the equilibrium η value for the inventory, η_{eq}. It is seen that $\eta_{eq} = 1 - D$.

The Salvatores analysis does not include losses to structural materials or parasitic capture to entrained fission products. We have attempted to incorporate these features into our analysis. In order to do this we have studied the chemical processing assumptions of the liquid-fuel reactors of the past and have developed models for neutron transport in reactor systems. We believe that discussion of this analysis will give the reader an understanding of the relationships between chemical processing, reactivity and accelerator beam current in actinide-burning systems.

The practical use of thermal systems depends on continuous or semi-continuous chemical processing. This idea is best illustrated by a discussion of the thorium fuel cycle that was extensively studied by Oak Ridge years ago, appearing in two different proposed reactor designs, the Molten Salt Breeder Reactor (MSBR)

(Robertson et al., 1971) and the Denatured Molten Salt Reactor (DMSR) (Engel et al., 1980). In the two studies, two completely different sets of fission product and ^{233}Pa decay assumptions were made. In a thermal system, thorium captures to ^{233}Th, decaying rapidly to ^{233}Pa, which has a half-life of 27 days. In a low-flux system, or if the ^{233}Pa is held external to the system, the decay is to ^{233}U, a fissile species. The net effect is that the thorium nucleus ultimately fissions after absorbing 2 neutrons. Because the number of neutrons released per fission is about 2.45, there is a net neutron gain in a well-engineered system. If there is significant capture in ^{233}Pa, the gain disappears. The reference MSBR design included a 2250 MW$_{th}$ reactor system with extensive fission product removal processing. Protactinium was continuously (3-day cycle time) recovered, allowed to decay to ^{233}U, and returned to the salt; approximately 6% of the ^{233}U was stockpiled as yield, giving a 1.063 breeding ratio.

Even though the chemical processing assumptions were stressing, the breeding ratio was small, leading to long doubling times. Because there were doubts whether the system would ever breed in practice with a realistic fission product removal system in place, a decision was made to cancel the program. The goal of the DMSR design was to provide a nonproliferation alternative system. The DMSR required continuous fueling with 20% enriched (^{235}U) and required minimal fission product recovery processing. No fissile material was recovered and ^{238}U was continuously added to satisfy the denaturing criteria; protactinium was not held external to the reactor.

The MSBR fission product recovery processing scheme has been examined and is presented in Table 3. The removal times are listed by chemical type. The rare earth removal times are the most stressing, and were never achieved on any large scale in practice. For more discussion, the reader is directed to Bowman (1992) or Robertson (1971). The impacts of breeding ratio (BR) and Pa cycle time were examined for the MSBR in the context that a transmuter employing thorium would be operated at an equilibrium breeding ratio of 1.0. Here we have defined "breeding ratio" as 1+ the rate that ^{233}U is removed from the system per fission. We adopted the DMSR treatment for protactinium: it is not held external to the blanket.

Table 3. Molten Salt Breeder Reactor (MSBR)
Fission Product Removal Processing

Group	Elements	Process	Removal Time
Rare Earths	Y, La, Ce, Pr, Nd, Pm, Sm, Gd	MSBR*	50 d
Rare Earth	Eu	MSBR*	500 d
Seminoble Metals	Zr, Cd, In, Sn	He Sparging	20 s
Noble Metals	Se, Nb, Mo, Tc, Ru,Rh, Pd, Ag, Sb, Te	MSBR*	200 d
Noble Gases	Kr, Xe	He Sparging	20 s
Volatile Fluorides	Br, I	MSBR*	60 d
U233 Precursor	Pa	MSBR*	3 d
Higher Actinides	Np, Pu	?	16 y

* - fluoridation-reductive extraction and metal-transfer process

The largest impact on reactivity comes from the parasitic absorption in the "non-fuel" actinides and the entrained fission products. These effects were calculated with ORIGEN2 (Croff, 1983) using a constant ^{232}Th feed and flux level of $1x10^{14}$ n/cm^2-s. The ORIGEN2 actinide and fission product one-group cross-section libraries were collapsed using the unit cell flux spectra calculated with MCNP (Breismeister et al., 1993) and the MSBR processing scheme described above. The results for the time-dependent ORIGEN2 calculations are presented in Figure 7 for the actinides only and for the sum of the actinides and fission products. The parasitic fission product impact on reactivity is small with the MSBR processing which essentially recovers all major fission product absorbers. Its value at equilibrium, the difference between these two curves, is $\Delta\rho_{FP} = -0.02$. The result is that the MSBR breeding ratio of 1.063 has been translated into a net reactivity margin of $\Delta\rho = (1.063 - 1.0)/\nu = +0.0257$. If the value of $\Delta\rho_{FP}$ were –0.0457 or worse the model reactor would not maintain criticality.

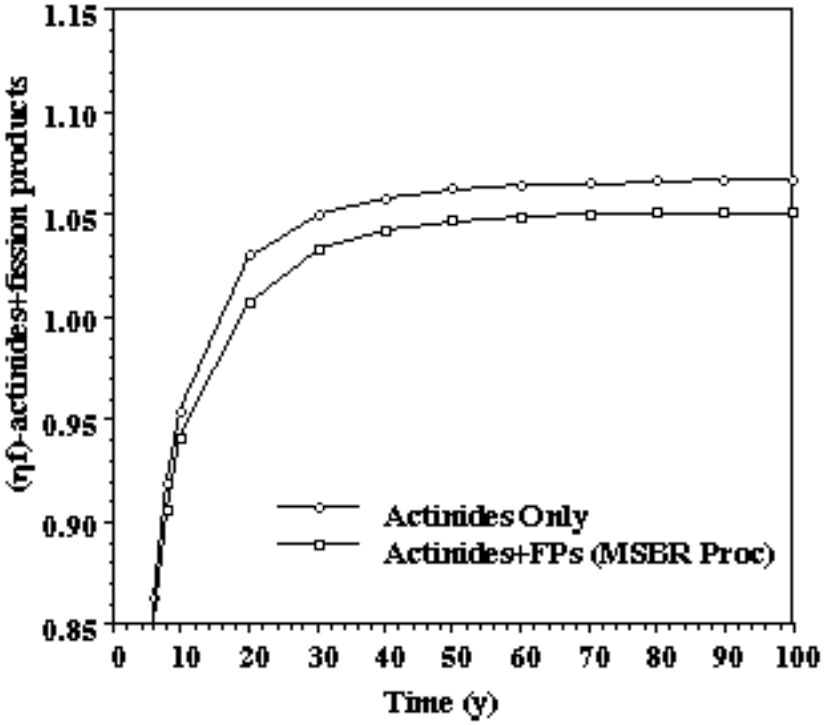

Fig. 7. Time dependent ηf value of actinides when combined with entrained fission products. The product of η and f is sometimes called the k-infinity of the fuel. The difference between the curves at equilibrium is 0.02.

If there is a spallation source provided, however, the system can operate regardless. Neglecting any changes in system leakage as a result of placing a target within the system boundaries, the relationship between beam current and the source's reactivity insertion $\Delta\rho$ is one of direct proportionality. This relationship can be derived by elementary principles from the fact that the system is at steady state. By calculating the reactivity lost to entrained fission products, the required beam current to operate versus residence time can be calculated. Of the MSBR processing assumptions, we believe that the most stressing one will be the 50-day residence time for the rare earth elements (minus Eu). Therefore, we choose this residence time as an independent parameter. In Figure 8 the required beam current to maintain steady state is shown as a function of this parameter. The processing times for other elements are held constant, with the result that no beam is needed unless the parameter is greater than about 200 days. The result is scaled so that 1363 moles/yr of fuel are being fissioned. It is assumed that the neutron yield from the spallation source is 40 neutrons/proton (corresponding to our estimates for a 1.6 GeV proton beam incident on a

molten lead target). This is an ideal situation where e. g., no additional capture reactions on impurities in the graphite are included. Such additional losses would be compensated for by additional current.

It is concluded that the accelerator can make a system feasible that previously may not have been. The accelerator beam is used to overcome the fission product burden from rare earth elements that may be difficult to remove continuously by chemical processing.

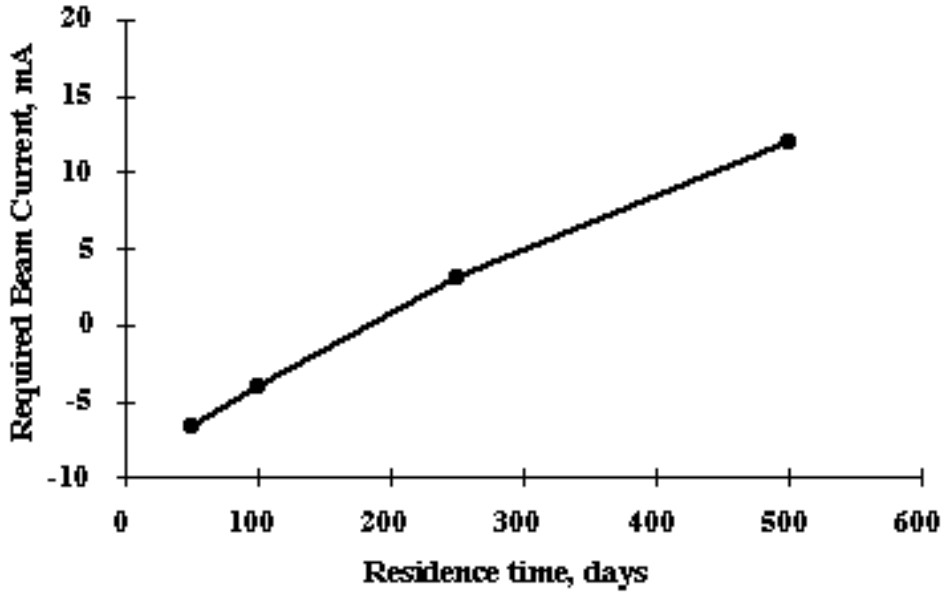

Fig. 8. Beam current required to maintain operation of a thorium/uranium reactor, based on the MSBR, with relaxed chemical processing assumptions for the rare earth elements. As these absorbers are left in the system for longer periods, more spallation neutrons are needed to maintain the system at steady state.

The reactor component of an ATW system has a similar neutron economy to the above MSBR-variant reactor. We have for this paper performed a simulation of this actinide-burning reactor at steady state. We generate results based on the coupling of whole-blanket calculations to equilibrium calculations. To find the equilibrium concentrations for the actinide mixture, we solved a set of coupled linear algebraic equations implied by steady state conditions using a code TRANEQI[2]. The inputs to the code are the 1-group spectrum averaged cross sections for all the reactions, the decay constants and branching ratios, and the total flux. The 1-group cross sections are calculated with the convenient tallying features of the established Monte Carlo neutron transport code MCNP.

The feed for the reactor is the sum of the transuranic species in LWR spent fuel discharge, which is 51% ^{239}Pu and 36% other plutonium isotopes. For the actinides, an individual nucleus may undergo fast or thermal fission or it may undergo a series of capture and/or decay reactions before ultimately undergoing neutron-induced fission. Because of the low capture-to-fission ratio for the feed, it is easy to imagine that an (initially) critical system can be built. After several years, in thermal systems in particular, the composition of the actinides within the transmuter approaches the "equilibrium" values, which in general result in a lowered reactivity. Detailed calculations have showed that the plutonium isotopes reach equilibrium after about 5 - 6 years while the curium isotopes take three times as long. The equilibrium average capture-to-fission ratio, however, is established when the plutonium isotopes are in equilibrium.

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[2]For each species the feed rate plus production from capture in and decay of other species is equal to the loss from fission plus capture plus decay.

The layout of the reactor model is shown in Figure 9. It has a heterogeneous lattice of molten salt fuel moderated with graphite. Above and below the lattice regions are plenums to transport the fuel to and from a heat exchanger that is located in the reflector.

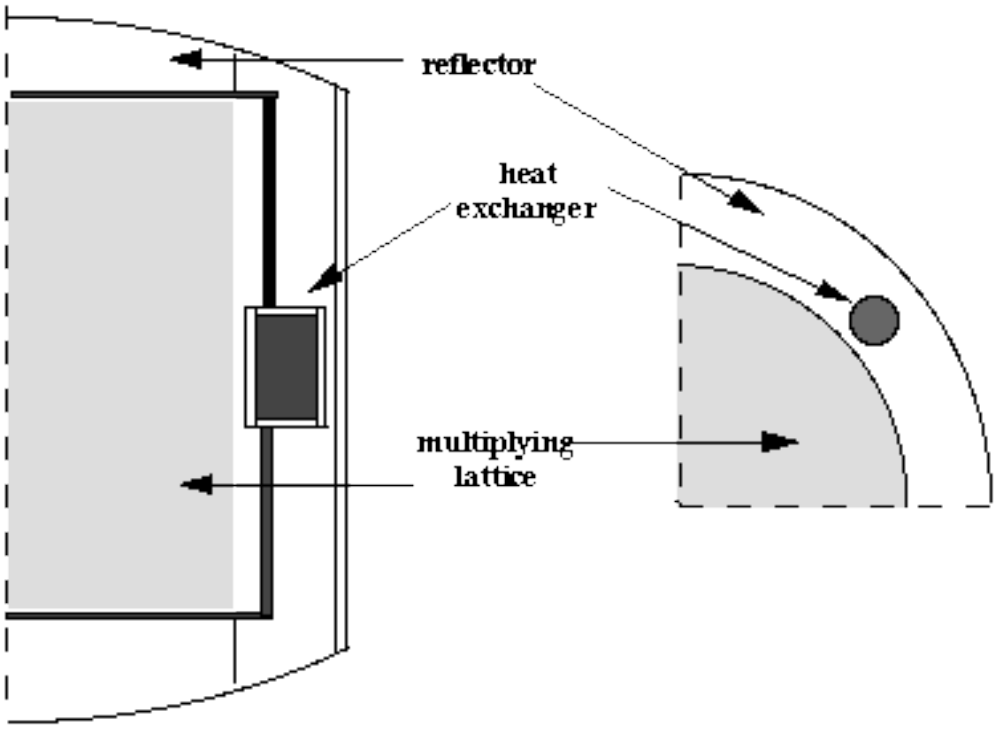

Fig. 9. Side and top views of the reactor model used for the neutronics calculations for the non-aqueous actinide burning system.

Because of thermal-hydraulic limitations and other design constraints, we have selected one design as a "baseline", to which we add departures. The baseline has a fuel power density of 400 W/cc in the fuel channels, a uniform temperature of 800 K, and a fuel channel diameter of 2 cm. The pitch between channels is 6 cm. The concentration of actinides is 300 grams/liter stated as AcF_4, which gives an equilibrium plutonium/salt mole ratio of ~1.3%, which is within solubility limits. The volume fraction of fuel in the heterogeneous lattice is 10%. Because the fuel is cooled in a heat exchanger, it spends about half its time outside the active region of the core. There are 6.4 m^3 of fuel in the lattice and 1.2 m^3 in the two plenums combined. The total power is 2560 MW_{th}.

The calculated k_{eff} for this baseline system is 1.0254, neglecting the effects of neutron capture on entrained fission products. As was discussed in some detail above, we estimate this *parasitic* capture will consume about 0.02 of the margin under nearly ideal (MSBR processing) circumstances. If k_{eff} is greater than unity the reactor can assist in LLFP transmutation; if k_{eff} is less than unity a neutron source is required. If the reactor is exactly critical, the actinide-burning portion of the system cannot contribute to LLFP transmutation and would not require a neutron source.

In Figure 10, results from some departures from the baseline design are given in order to show what kind of options are available to increase performance. In the top-most curve the effect of increasing the power density is given. In general, the higher the power density, the lower the equilibrium capture/fission ratio for the actinides because of the increased fissioning probability of short-lived species such as ^{238}Np (Bowman et al., 1992). In the middle curve the fuel volume fraction in the heterogeneous matrix was varied by increasing or decreasing the diameter of the fuel channels. If the volume fraction is too low, there is excessive parasitic capture in the graphite. The bottom curve shows the effects of changing the moderator composition and temperature. The presence of beryllium changes three things. Parasitic capture increases, but this is compensated for by an increase in gain from (n,2n) reactions. Beryllium also

achieves a cooler neutron temperature than graphite, resulting in a better equilibrium capture/fission ratio for the actinides. The lower-temperature systems would require a thermal decoupling between the moderator and the fuel, possibly by the introduction of an insulating region. There would be a fairly dramatic improvement in performance if the engineering details could be worked out.

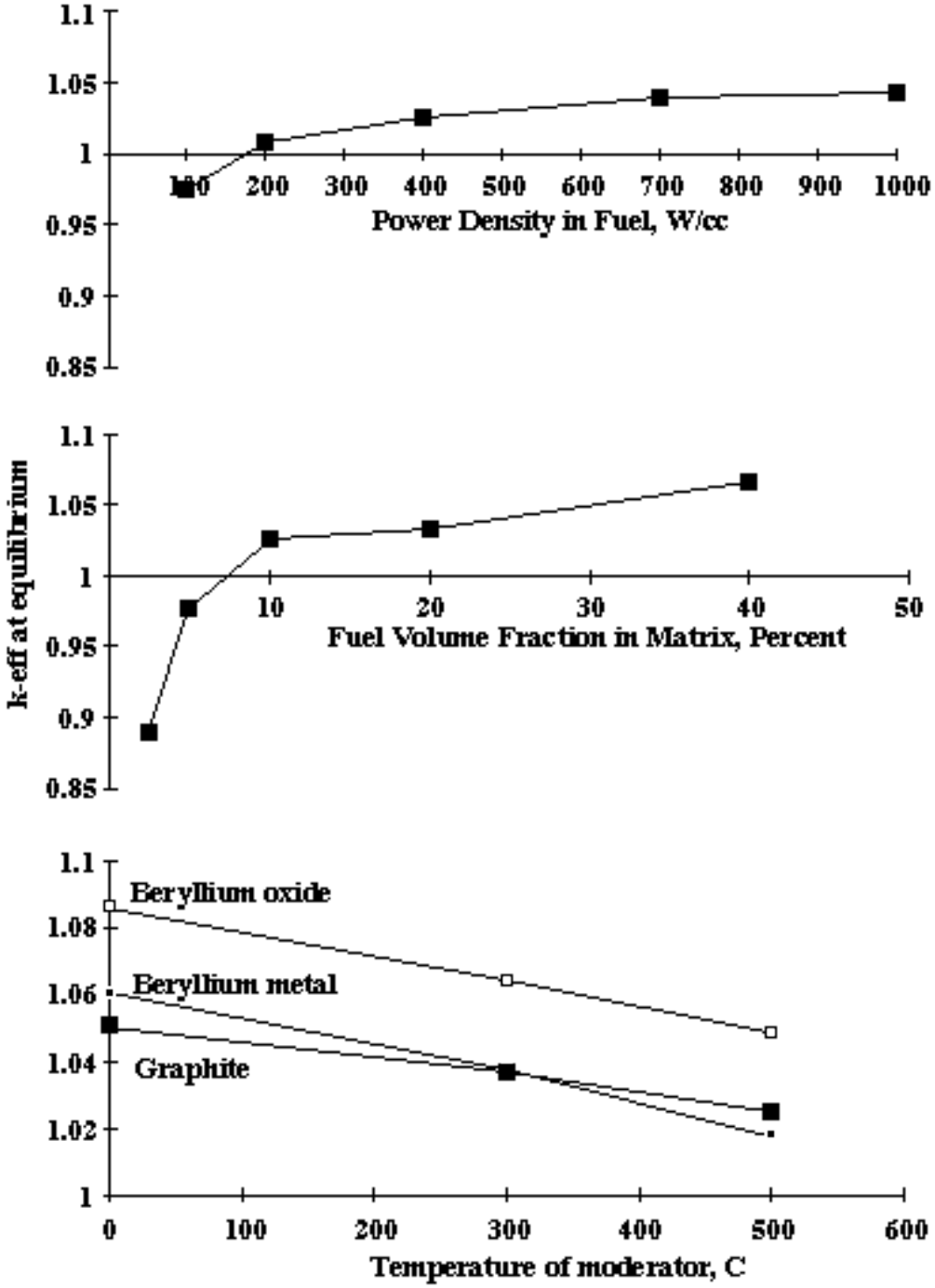

Fig. 10. The effects of various model design changes on the keff of the LWR-discharge burning reactor. The "baseline" design is graphite moderated, has a 10% fuel volume fraction, and 400 W/cc power density in the fuel.
(top) Increasing the power density lowers the equilibrium capture/fission ratio in the actinides, increasing k_{eff} .
(middle) If the volume fraction is too low, graphite parasitic capture lowers k_{eff} .
(bottom) Changing the moderator to beryllium or beryllium oxide and/or lowering the moderator temperature improves k_{eff} .

Table 4 lists some information about the LLFPs. All are characterized by low β decay energies; some are accompanied by gamma rays. In the last column is a CANDU-spectrum one-group capture cross section, taken from the library provided with the ORIGEN2 code (Croff, 1983).

Table 4. Long-lived fission products (LLFPS) of potential interest in LWR spent fuel transmutation.

species	half life	beta energy	gamma energy	thermal cross section
	1000's yrs	keV	keV	barns
^{79}Se	65	160		1.5
^{93}Zr	1500	60		2.1
^{99}Tc	210	293.0		17
^{107}Pd	6500	33.0		6.6
^{126}Sn	100	250.0	87.0	0.16
^{129}I	16000	150.0	39.0	15
^{135}Cs	3000	210.0		5.8

In Table 5 the annual discharge molar quantities of each species found in LWR spent fuel (1 GW_e operating at 100% capacity factor 33,000 MWd/MTHM[3] burnup) are listed. The "isotopic moles" specifies the moles of the isotope if it is the only species to be transmuted. In order to transmute these species, one or more capture reactions on the nucleus leads it to where it can undergo β^- decay. Most species occur generally with several isotopes, as seen in Figure 11. Zirconium appears as 6 species, 90 through 96, not including the short-lived 97. If isotope separation is not used, isotopes 90 through 94 must be transmuted to 95, which decays by two successive β^- emissions to ^{95}Mo. The isotopes 90 through 93 thus require *more than* 1 neutron per transmutation. Isotopes 94 and 96 require each 1 neutron. The net average per elemental atom (not per atom of the long-lived isotope) is 2.1 neutrons. These are the quantities listed in the middle column in Table 5. Note that this number is 1 for iodine, because the two isotopes (127 and 129) each require only one neutron capture each. The last two columns give the number of neutrons consumed to transmute these species isotopically and elementally per actinide fission.

Table 5. Molar quantities and mole ratios to transmute the LLFP species.

species	isotopic moles	elemental moles	elemental neutrons required	isotopic neutrons per actinide fission,	elemental neutrons per actinide fission,
	per LWR-yr	per LWR-yr	per atom	dimensionless	dimensionless
^{79}Se	2.5	23.8	1.2	0.002	0.021
^{93}Zr	257.5	1297.4	2.1	0.189	2.000
^{99}Tc	259.5	259.5	1.0	0.190	0.190
^{107}Pd	67.9	427.3	2.8	0.050	0.878
^{126}Sn	7.2	46.0	1.8	0.005	0.061
^{129}I	46.2	60.5	1.0	0.034	0.044
^{135}Cs	74.1	588.1	1.6	0.054	0.690

[3]units are Megawatt-days per Metric Tonne Heavy Metal

Not included in any of the columns in this table are the internally-generated quantities of LLFP species which are produced by the fission of higher actinides. On average the internally generated quantities are 10% to 40% of the external burden, but can vary with the on-line chemical processing assumptions and flux level in the system. Calculations we have performed with ORIGEN2 (Beard et al., 1994b) have shown that for flux levels less than $2x10^{15}$ n/cm^2/sec the internal burden is independent of flux to within ±10%. In our generic calculations we have taken a highly thermal neutron energy spectrum and a 30-day mean residence time for fission products in the actinide burner.

The neutron balance and beam current of the ATW system in either the monolithic or the two-subsystem configuration is a strong function of the choices that are made for the burning of LLFPs, because most of the net neutron balance used to transmute LLFPs are ultimately supplied by the accelerator. This is a corollary to the calculational result that the reactor component of the system is critical or nearly critical at equilibrium, or to the conclusions reached by Salvatores et al. (1994).

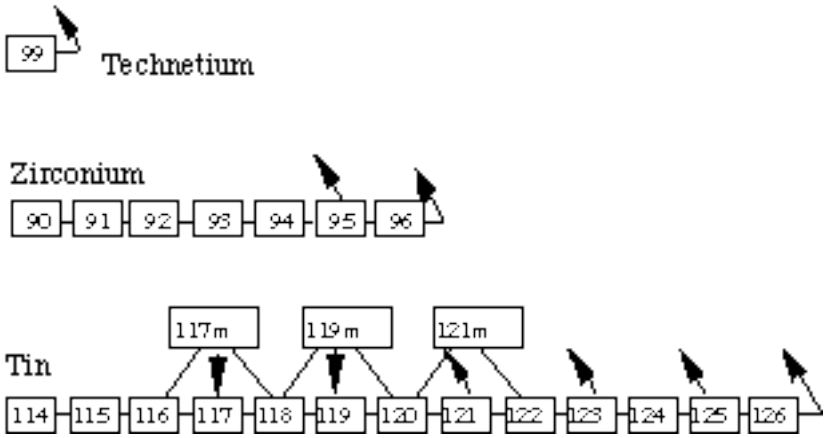

Fig. 11. Transmutation schemes for some long lived fission product species. Upwards arrows represent β- decay, which eliminates one atom of the element. Technetium is the only element of interest where a single neutron need be captured for transmutation. Others are more complicated, requiring more than 1 neutron per LLFP nucleus on average.

Using a simple neutron balance calculation, the proton beam power requirements for transmutation were calculated. It was *temporarily* assumed that no LLFP transmutation is provided by the reactor. Based on computational experience using the code LAHET (Prael and Lichtenstein, 1989), we select a reasonable number of neutrons liberated from the spallation target at c_{np} = 25 neutrons/proton per GeV of beam energy. The capacity factor is 100%. The target beam power P_i required to transmute a species i requiring α_i neutrons per actinide fission (as in the last two columns of Table 5) is then $P_i = \frac{R_A \alpha_i}{c_{np} c_o}$ where R_A is the rate of actinide burning = 1363 moles/year and c_o = 0.322 moles/year/mA (simply a unit conversion factor). The power required to *operate* the accelerator may be a factor of two or more greater (Lawrence et al., 1991) due to the various electrical conversion losses. Also not included in the calculation are any effects from neutron leakage or parasitic capture in the target, which will reduce the efficiency of the process. It is estimated from our computational experience that for the species with high thermal neutron capture cross sections (> 10 barns), the efficiency can be 65% - 80% or higher if a lead target is used. Alternatively, if the target material were constructed of, e. g., lead mixed with ^{126}Sn, some proton transmutation would occur without significant loss of neutron yield (Carter et al., 1994).

In the upper and lower parts of Figure 12 the amount of beam power provided by the accelerator is shown (on a per LWR basis) for each of the LLFPs, on an isotopic and elemental basis, respectively. Shown are

both the external and internal transmutation burdens. Transmutation of the only species that appears as a single isotope (^{99}Tc) would likely require ~80 MW of electrical power, when conversion factors and leakage are included. It is to be remembered that this calculation does not include any transmutation provided by the actinide-burning reactor. When isotopic separation is allowed, the transmutation of all 7 isotopes would require 89 MW of beam power, which scales to ~1/4 the power of the LWR when inefficiencies are considered. Without isotopic separation, the elemental ^{93}Zr by itself would require ~80% of the power produced by the LWR.

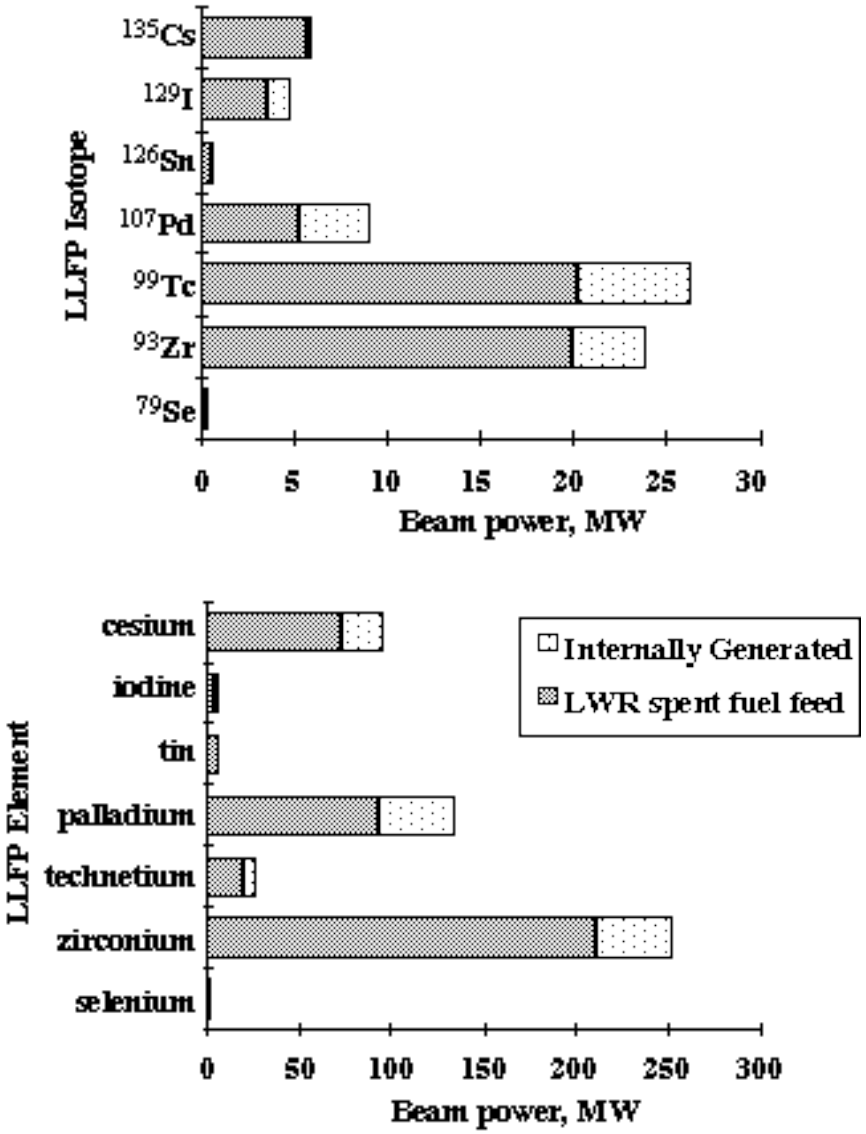

Fig. 12 (upper) The proton beam power at target for each LLFP isotope transmuted, when it is assumed that isotopic separation is used. (lower) The same, when isotopic separation is not used. There is no transmutation of LLFPs by the reactivity margin supplied by burning the actinides in this calculation. The power required to operate the accelerator will be higher because of conversion losses.

The reactor in the system can help to transmute LLFPs in two ways (1) it burns them in its reactivity margin and (2) it supplies electrical power to run the accelerator. Under the ideal (MSBR) chemical processing assumptions, it has a margin of $\Delta\rho = 0.0054$ for burning LLFPs that are placed in the heterogeneous lattice. The amount of LLFP transmutation per actinide fission provided by this margin is

$\alpha_{LLFP} = \nu\Delta\rho = 0.016$, where ν is the neutrons emitted per fission at equilibrium = ~3.05. Some additional capacity for LLFP transmutation is available in the leakage regions. The ratio of leakage to fission neutrons, α_{Leak}, is equal to 0.10 in the model reactor. To utilize these neutrons for transmutation, LLFP species would be placed in the outer portion of the reflector. Their presence does not affect the neutron balance otherwise. The maximum rate at which the reactor can transmute fission products, including the use of leakage neutrons is $\alpha_{LLFP,max} = 0.116$. For additional species transmuted beyond that provided by the reactor, the full incremental amount of power shown is required. The actinide-burner reactor can supply 340 MW of AC power (for a 40% efficient thermal/electrical conversion); the system could be a net producer of electricity even if all the isotopic species shown in Table 4 are transmuted.

By comparing the $\Delta\rho$ values used to transmute LLFPs with the margin lost to *parasitic* fission product capture, it becomes clear that the validity of these estimates depends heavily on the ability of the system to remove fission products.

Mass and Energy Flows For ATW System

In this and subsequent sections we give flow diagrams that present ATW system variations in terms of connected units. Our calculations here are scaled to systems that transmute a single 1 GW_e reactor-year of LWR transuranic waste per year of operation. We take this to mean 33 tonnes of uranium (MTU) discharge after 33,000 MWd/MTU burnup, which translates into 1363 moles, or 327 kg of transuranic species per year. Calculations are representative of a 100% capacity factor for the LWR and the transmutation systems. The balance is derived from the baseline reactor model. It is assumed that the reactor component is used to transmute LLFPs in its margin and with its leakage neutrons with $\alpha_{LLFP} = \alpha_{LLFP,max} = 0.116$. When the two systems are combined into one the number of neutrons available per fission may be slightly higher than this, but counteracting this is the fact that the efficiency for capturing the leakage neutrons will be less than unity.

The mass and power flows for the baseline ATW system, scaled appropriately, are shown in the upper third of Figure 13. In this specific example the flow rates correspond to the transmutation of the actinides and ^{99}Tc and ^{129}I. There is no isotopic separation used for these balances, therefore ^{127}I is burned also. Heat produced is used to generate steam that runs turbine/generators. The efficiency for conversion of electrical power to proton beam power is taken as 50%. The accelerator uses less electricity than is made, and the surplus is sold to the grid. A small facility removes LLFPs created by fission for recycle and continuously recycles the actinides. The transmutation products of the LLFP destruction are removed by this facility. There are a greater number of moles of transmutation products than of LWR LLFP feed because of internal generation of LLFP species.

The flow diagram is shown again in simplified form in the middle of Figure 13. The basic module of an accelerator-based system is a box that consumes 1 LWR worth of higher actinide discharge, and 1 LWR worth of LLFP discharge. The module's outputs are the LLFP transmutation products, the other fission products, and a certain amount of electricity. Not shown are at least three items which may be of importance: the waste heat (equal to the fission heat plus LLFP transmutation heat minus the electrical output), the activation product stream (Bezdecny et al., 1994), and spallation product stream. It is believed that the activation stream will depend heavily on the detail design and materials of construction of the system components, but that in general none of the species listed in Table 4 will be produced. An exception may be if zirconium or nickel alloys are used in construction, producing ^{93}Zr and ^{59}Ni, respectively.

The system is shown again as two subsystems that accomplish the same goals in the bottom third of the figure**.** The actinide-burning task is performed in a reactor that is fueled with the actinides. Electricity produced in this reactor is sold to the grid. Only a portion of the LLFP feed is transmuted with the

actinides, and the rest is sent to an accelerator-based system for destruction by neutron capture. The LLFPs generated by the fission of the actinides are added to this stream. There is no electrical production in this portion of the system, and the heat generated is dumped. The accelerator buys electricity from the grid.

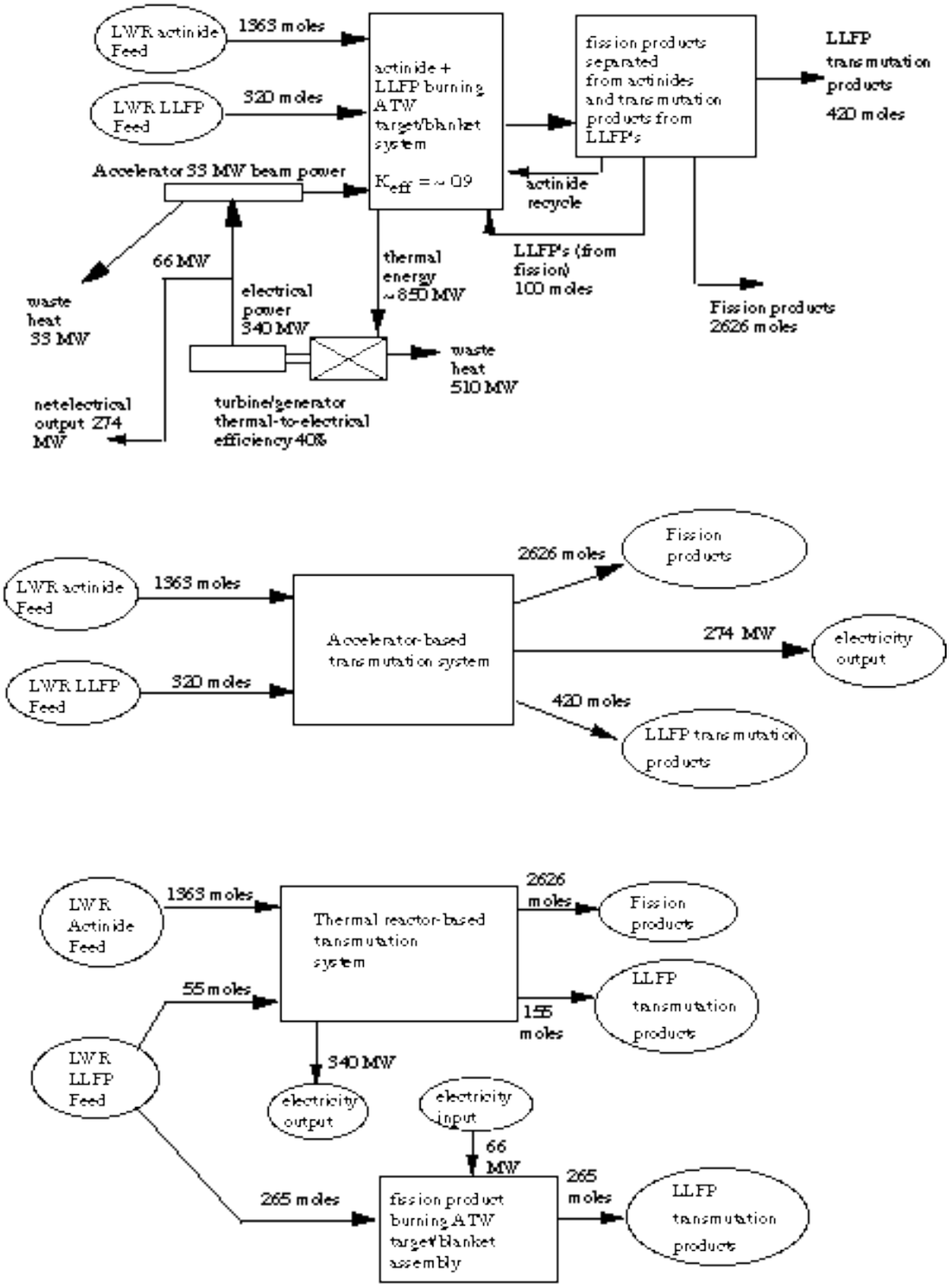

Fig. 13. (upper) Baseline ATW system scaled to 1 LWR worth of actinide and LLFP waste from commercial LWR spent fuel. The LLFPs are technetium and iodine. (middle) Stated as a "black box" that transmutes 1 LWR worth of higher actinides

and 1 LWR worth of LLFPs, burning the LLFPs that it makes from fission.
Electrical output of 274 MW (net) is produced.
(bottom) Decomposed in terms of mass and energy flows into a reactor-based system and a LLFP-burning ATW target/blanket assembly.

The actinide-burning task is assigned to a reactor system that necessarily contains other components such as a chemical processing plant to remove fission products (in the case of a thermal spectrum system), a steam supply system and a turbine/generator system. The neutron balance for either configuration is taken to be the same, but this neglects leakage in the spallation target/blanket in the lower configuration. Another difference in the net energy streams between the two configurations is that the beam power is recycled to the thermal system in the first case, but the beam power is a small fraction of the total power.

Addition of HEU Feed into ATW systems

The addition of a fissile species into the fuel cycle can increase the reactivity margin of the "reactor" component of transmutation system, as shown in Figure 14. The margin is then used to burn more of the LWR discharge LLFPs, resulting in a lowered beam current. The (93%) highly-enriched uranium (HEU) component of the system has a high η_{eq} value, making it potentially very useful for this purpose. The ratio of fission power in each "reactor" can be arbitrarily varied. In this case the ratio is 1:1, resulting in a doubling of the system fission power (versus no HEU use), giving more than twice as much net electrical output and a smaller size of accelerator. Whether or not this is more advantageous depends on viewpoint, i. e., the same amount of nuclear waste is being destroyed per unit time in a more energy-efficient system that may have higher capital costs. The proliferation dangers of HEU use may preclude this option commercially, although HEU fuel is routinely used in naval reactors.

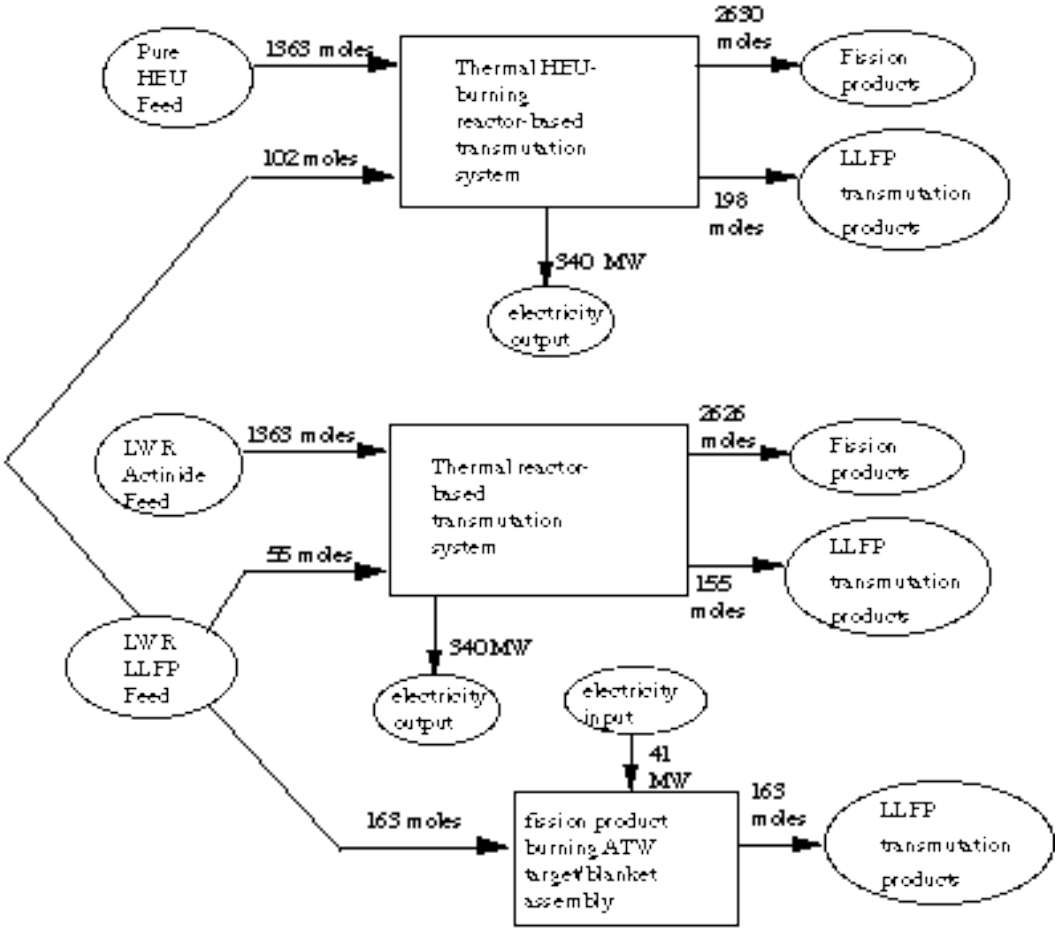

Fig. 14. HEU-FED ATW system scaled to 1 LWR worth of actinide and LLFP waste

from commercial LWR spent fuel. The LLFP burning is the same as before. The fission power from the HEU component is equal to the fission power from burning the LWR-discharge actinides in this case.

It is estimated that the U. S. stockpile may contain 500 tonnes HEU and the former Soviet Union stockpile may contain up to 1400 tonnes HEU (Marias et al., 1993). There have been discussions of releasing 500 or more tonnes of this supply into the world marketplace. It is planned to be released in the form of commercial-grade fuel of ~4% enrichment after blending with natural or depleted uranium. If 600 tonnes were instead used in a transmutation scheme like that shown in Figure 14, with a 1:1 feed ratio to LWR discharge actinides, it would be sufficient to transmute all the spent fuel currently stored in the United States.

Addition of Thorium Feed into ATW systems

The known reserve of thorium in the US is so large that it could supply the fuel for the current world nuclear capacity (325 GW_e) for the next 2200 years (Benedict et al., 1981). If thorium is to assist in transmutation, it must burn its own LLFPs generated by fission internal to the system, so it must have $\alpha_{LLFP} = 0.071$ (the Tc and I yields are about 5% less than for plutonium fission), requiring $\Delta\rho = 0.0289$. The margin in the model thorium reactor is $\Delta\rho = 0.0257$. Because of the slight deficit, thorium cannot be said to help significantly in transmutation, although it may be able to burn its own fission products if it uses leakage neutrons. The balance reflecting this conclusion is shown in Figure 15. Modifications to the moderator composition and/or temperature that were discussed for the LWR-discharge burning reactor would apply to this analysis as well, possibly changing this balance favorably.

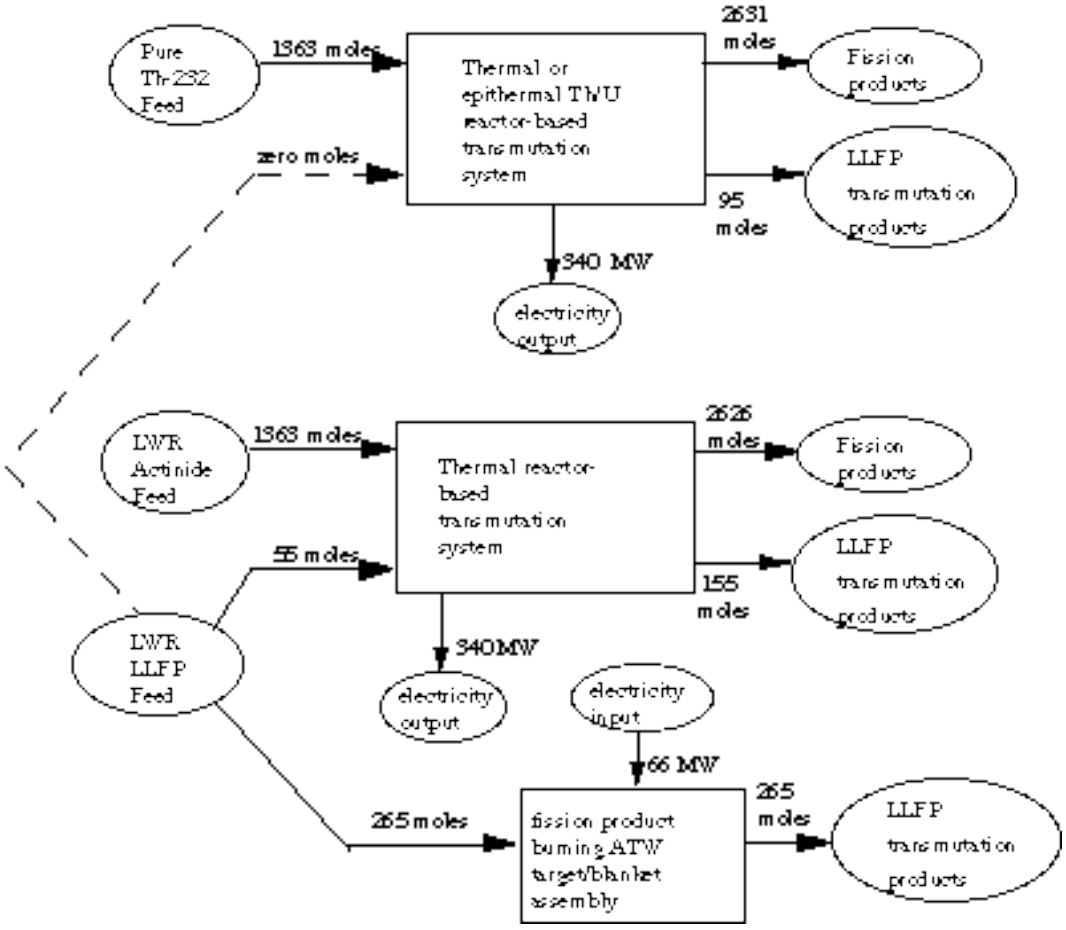

Fig. 15. Thorium-fed ATW system scaled to 1 LWR worth of actinide and LLFP waste from commercial LWR spent fuel. Some additional LLFPs are generated

within the thorium "reactor", but burned internally. There is not enough of a margin within this reactor to burn LLFPs from outside. This combined system could not only burn LWR waste but replace the power lost when the LWR retires.

A thorium-burning reactor or accelerator-driven system, if engineering practicalities were overcome, could continue to provide replacement nuclear electricity for retiring LWR's without generating any transuranic high-level waste stream or long-lived ^{99}Tc or ^{129}I. If it proved impossible to build it as a critical system because of fission product removal rate limitations, the neutron balance could be maintained with an accelerator-driven spallation source (Venneri, et al., 1993; Aldhous, 1993).

Comparison of ATW Reactor Component with ALMR

If the ATW system is to be physically divided into a parallel reactor and LLFP-burning accelerator subsystems, there is no longer a strong constraint that the reactor component employ a neutron energy spectrum that is useful for LLFP burning, and a fast reactor becomes an option.

A fast spectrum gives a lower capture-to-fission ratio for the actinides, resulting in a much improved neutron economy in theory. However, in most practical designs the excess neutrons are lost to control rods and leakage and cannot be used for transmutation. With re-designing, some of the fast reactor's reactivity margin could be used for burning LLFPs which would improve the efficiency of the overall transmutation system. The design and testing of subassemblies for burning LLFPs in fast reactors has been performed (Salvatores et al., 1993; Wootan and Jordheim, 1991).

The burning of actinides in any solid-fueled reactor is limited by loss of doppler feedback when the resonance absorber is eliminated. Therefore, safety and transmutation are traded against one another. Replacement of the standard resonance absorber, ^{238}U, with a non-actinide could provide a way around this problem (Wootan and Nelson, 1993). Alternatively, a fluid-fueled fast reactor would have the advantage that the prompt temperature coefficient is provided by thermal expansion of the fuel. This would allow continual fission product removal for reprocessing and removal of transmutation products. Power control by an accelerator could provide a degree of control and safety for higher-actinide burning fast systems not available in conventional reactors (Rief and Takahashi, 1994).

Direct economic comparison between the molten-salt reactor and the ALMR has not been performed. Parametric scoping studies have shown that the cost of electricity from the ATW reactor component is likely to be higher than LWR systems mostly because of the chemical processing equipment capital costs (Krakowski, 1994), but the ALMR would also have these type of costs.

Fast reactors have the drawback of high in-core actinide inventory per unit burn rate. The model of Pigford and Choi (Pigford and Choi, 1991) in the past has been used to compare various alternative processing and transmutation schemes for LWR spent fuel waste, e. g., Ramspott, et al., (1992). It compares the transmutation capabilities of systems based on an inventory reduction factor that is computed based on all the transuranic species present including unprocessed spent fuel, transmutation system internal inventories and transuranic species present in waste streams. Because of this last consideration it is important to use a realistic decontamination factor, γ, in the calculations. We have chosen $\gamma = 1000$, which corresponds to a 0.1% loss in processing, both in the front end and in refueling of ALMR cores and the on-line processing in the ATW reactor component. Adaptation of the model to ATW is explained in Davidson (1992). No credit is given for destruction of LLFP species in this model. The results of the calculation are stated in terms of time-dependent inventory reduction factor $\psi(t)$, which is defined as

$$\psi(t) = \frac{\text{(time dependent inventory of TRU in unreprocessed LWR spent fuel waste without actinide burning)}}{\text{---}}$$

(time-dependent inventory of TRU in the actinide-burning reactor and its fuel cycle and in the high-level waste from reprocessing LWR and ALMR fuel)

All required system and high-level waste inventories of TRU are included. In general, the larger the inventory reduction factor the better, provided that it is achieved on a reasonable time scale. The results of the calculations are shown in Figure 16. The ALMR curve is the same as is shown in Figure 1 of Pigford and Choi (1991). Different breeding ratios can be used for the ALMR, but $\beta = 0.62$ is the value preferred by the U. S. Dept. of Energy, according to this reference. For further explanation of the model derivation the reader is directed to that paper and references therein. The results show a clear advantage for the ATW reactor component subsystem on time scales of a century or less. On this basis, the transuranic burning capability of the ATW concept appears to be quite competitive versus the ALMR.

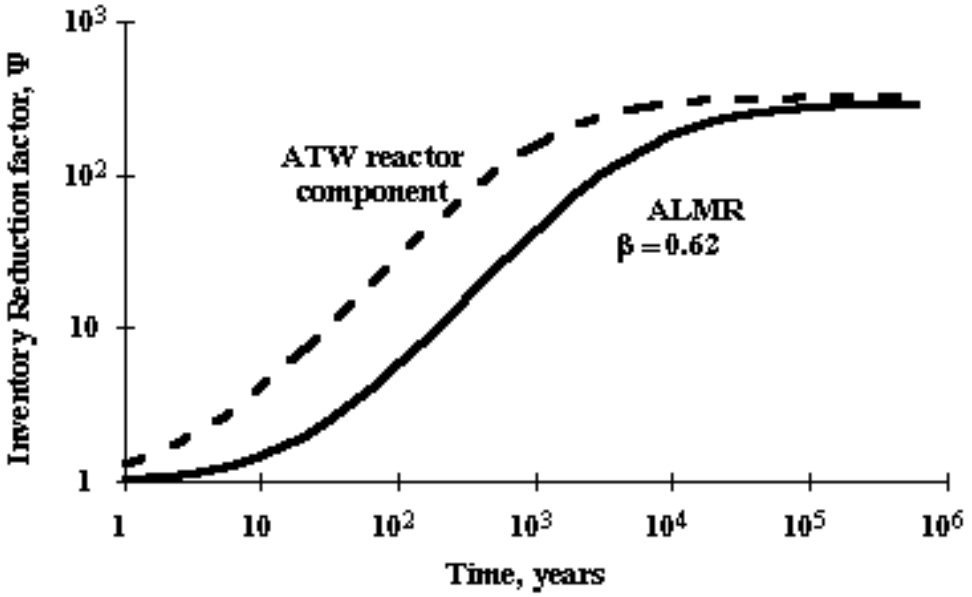

Fig. 16. Comparison of the ATW's actinide transmutation capability versus the ALMR ($\beta = 0.62$) according to the Pigford-Choi model. An inventory reduction factor of 10 is reached in 30 years for the ATW, while it takes the ALMR 200 years to reach this factor.

Conclusions

Although the main justification for transmutation has been in the potential economic benefits from fissioning the transuranic species, if it is to improve the prospects for dealing with high-level waste problems it will also need to transmute some of the LLFPs. The methodology discussed in this paper provides a simple way to estimate the electrical requirements for the accelerator transmutation of each LLFP species. If the actinide-burning reactor transmutes LLFPs in its margin the economy improves. The addition of highly enriched uranium feed would increase this benefit.

The configuration of an ATW system as a separate thermal actinide-burner in parallel with an LLFP-burning spallation target/blanket may be worth consideration because of possible operational advantages. Aside from small leakage differences, the neutron balance is unchanged versus a monolithic configuration that accomplishes these goals. It is found that the thermal actinide-burner achieves a more rapid actinide inventory reduction than an ALMR. This actinide-burner can alternatively be operated as an accelerator-driven subcritical system with relaxed chemical processing requirements.

The US General Accounting Office issued a report emphasizing the negative overall technical conclusions of the Ramspott, et al. study, adding that the development of transmutation technology into a full-sized

productive operation would take decades and cost billions of dollars (GAO, 1994). Their final conclusions were positive, however, about the usefulness of transmutation under certain circumstances "If nuclear power continues to be used in the United States and if waste transmutation could be proved technically and economically feasible, not as much long-lived radioactive waste would be produced if future power needs are met by using a new generation of power producers that are designed to economically burn their own waste." It is because of these potential circumstances that we have discussed the thorium cycle.

If the US were to remain committed to nuclear power with the once-through fuel cycle for LWR's, large repositories would have to be constructed every one or two decades. The thorium-reactor system, possibly driven by an accelerator, could provide replacement capacity for retiring LWR's without the continuing geological waste disposal problem. Transmutation of existing waste is a concept that provides a bridge between the current state of affairs where waste is accumulating and not being disposed of, and a future where a few small repositories or storage facilities could hold the waste generated by many power stations in a way that is acceptable to the public.

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